PRESERVATIVE LEACHING FROM CHROMATED COPPER ARSENATE (CCA) TREATED TIMBER TO THE SOIL ENVIRONMENT

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ABSTRACT

Wood has been used for centuries as building materials and the concern of leaching of toxic elements into the soils and groundwater arise when Chromated Copper Arsenate (CCA) wood preservatives were used to extend the service life of timbers in soil contact against decay and termites infestation. In this study, the levels of total copper, chromium and arsenic within CCA-treated poles and in soils surrounding the poles in service at a plot in Timber Research and Technical Training Centre, Kuching, Sarawak, Malaysia since 1980 or 1981, were investigated. There are significant decreases of CCA salt retention in poles after 30 years of weathering (p<0.05) but with insignificant differences (p>0.05) between retention of CCA salt in wood 1300 mm above ground level and 0-20 mm below ground level. The levels of these elements were found to be significantly elevated compared to control sites (background level) in soils immediately adjacent to the treated poles. The metal levels were highest at the soil surface in contact with the poles but decreased with distance and depth. Mean extractable arsenic levels ranged from 14.53 up to 100.06 mg/kg, chromium levels from 23.31 to 148.32 mg/kg and mean copper levels from 21.83 to 104.72 mg/kg. The results show that the soil is contaminated, albeit slightly, after 30 years exposure of CCA treated wood in ground contact.

Keywords: Treated wood, chromium, arsenic, copper, CCA wood preservatives, soil contamination.
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<td>CCA</td>
<td>Chromated Copper Arsenate</td>
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<td>AR</td>
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1.0 INTRODUCTION

The unique characteristics and versatility of wood makes it an extensively used natural resource worldwide. Conservation pressures and scarcity in naturally durable timber supplies have led to increase in usage of treated timbers worldwide. Timber from sustainable forests will continue to provide economically abundant structural building materials and wood products. However, wood is a natural biological material and will be subjected to decay and infestation by many biological organisms and degradation by weathering elements. In order to extend the service life of wood, preservatives are used to make it toxic for wood-degrading organisms. In ground contact, CCA preservative treated wood with high retention values may last 10 times longer than untreated wood.

In Malaysia, chromate copper arsenate (CCA) are commonly used to treat wood for structural uses like timber roof trusses under Hazard class H2 (Wong et al., 2006) and utility poles. However, leaching from such treated wood of heavy metals has led to CCA treated timbers to be subjected to stringent environmental controls in many industrialized countries where uses of heavy metals in wood preservatives have been slowly phased out in recent years in preference for organic active ingredients instead (Cooper, 1994; Lebow, 1996; Hingston et al., 2001; Wong et al., 2006).

Wood preservative treatments used vary according to the type of active chemicals employed. Some of the examples are Alkaline copper quarternary, Creosote, Copper Azole, Light Organic
Solvent Preservatives (LOSP), Copper HDO and Chromated Copper Arsenate (CCA). The wood preservative used of interest in this paper is CCA.

CCA treatment is claimed to provide stable deposition of wood preservatives in the wood when done correctly but in reality leaching of CCA components will occur due to weathering effect over time and many researches have been conducted to answer several key questions on how such leaching can occur. However, more questions arise due to lack of data on the migration of CCA components from treated wood into the soil and only recently when researchers are actively looking into this. Morrell and Huffman (2004) did a comprehensive study on the metal levels in soils surrounding CCA treated timber in Florida (a sub-tropical region) but there is lack of data concerning high humidity and rainfall tropical regions such as Malaysia. This is a study to fill in those gaps of knowledge.

The objective of this study is to determine if there is long term CCA preservative leaching from treated Malaysian hardwood poles into the surrounding soil, by assessing metal levels of wood preservative precursor (Chromium, Arsenic, and Copper) in soil surrounding CCA treated poles as well assess residual CCA retention at aboveground and below ground line in treated wooden posts.
2.0 LITERATURE REVIEW

2.1 Chromated Copper Arsenate (CCA) treatment

Chromated Copper Arsenate (CCA) is one of wood preservatives used to treat and increase service life of wood. Undeniably, CCA is among the most effective in protecting wood from a wide spectrum of wood degrading organisms mainly decay fungi and insects (Wong et al., 2006; Wong and Lai, 2007). Furthermore, CCA is also popular due to its cost competitiveness. In fact, CCA treated timbers are used widely in landscape, gardens, structural application and many other outdoor applications in United States, Australia and Britain. Furthermore, CCA treatment makes up 75% of total volume and 97% of the waterborne preservative market in the US (Solo-Gabriele et al., 1999).

Furthermore, CCA has dominated the treated wood market for several decades and only recently CCA usage is slowly being phased out mainly in the developed economy regions due to concerns with the use of arsenic as one of its active ingredient. The potential of environmental hazards had lead to current established CCA treated wood market to restrict the use of CCA treated timbers to non residential environments and promote the use of relatively less effective environmentally preferred organic wood preservatives (Cooper, 1994; Lebow, 1996; Hingston et al., 2001; Stilwell et al., 2003; Wong et al., 2006; Wong and Lai, 2007).
Each component in CCA has its own function and purpose. Chromium serves as the catalytic anchor which fixes the two elements, arsenic and copper that acts as insecticides and fungicides respectively to the cellulose and other wood elements (Dawson et al., 1991; Chirenje et al., 2003). Reduction reaction of chromium is important to properly serve its purpose as anchor of arsenic and copper. Indeed, as CCA are considered “fixed” when chromium are reduced from chromium (VI) to chromium (III) species and many authors are investigating this fixation reaction chemistry and leaching performances especially at temperate countries (Cooper and Ung, 1992; Wong et al., 2006; Salim et al., 2012).

Retention of CCA salts or oxide in wood is controlled via specific industrial treatment schedules, depending on the purpose of usage and also wood properties. The retention values range from 4 to 40 kg/m³ with above ground application at 4 kg/m³, structural pole at 9.6 kg/m³, freshwater contact at 6.4 kg/m³ and saltwater application is between 12.8 to 40 kg/m³ (Chirenjea et al., 2003).

2.2 Leaching of CCA Components

2.2.1 Issues of leaching

In recent years, environmental concerns and exposure to CCA treated wood may present certain hazards with regard to the use and disposal of CCA treated timbers where it may come into direct or indirect contact with water or human. Most of the criticism center on the use of arsenic as one of the key ingredients of the preservatives.
Many authors report different results of leaching rates of CCA to the soil environment. Townsend et al. (2000) determined the concentration of arsenic, chromium and copper from CCA treated timber decks in Miami, Florida to be 28, 34 and 40 mg/kg compared to background level of 1.5, 10 and 10 mg/kg respectively. Studies conducted by Morrell and Huffman (2004), determined that the mean extractable arsenic adjacent to the pole ranged from 1.74 to 8.19 mg/kg with high background concentration at 0.38 mg/kg while further distance away at 300 mm from pole, the mean arsenic level is found to decline to levels ranging from 0.06 to 0.80 mg/kg. Besides arsenic, they reported copper leaching to be in range of 15.8 to 301.0 mg/kg adjacent to treated pole and declined to a mean range of <0.1 to 5.9 mg/kg furthest from the pole (at 300 mm) while chromium was found to be at a low level of 1.02 to 0.27 mg/kg next to the pole and declined close to background levels of <0.2 mg/kg and ranging from 0.02 to 0.06 mg/kg.

Utility poles treated with CCA are partially buried into the ground and when leaching of CCA components occur, soils surrounding CCA treated timbers are found to be elevated in arsenic, chromium and copper (Townsend et al., 2003; Morrell and Huffman, 2004; Mercer et al., 2012). In fact, As, Cu and Cr concentrations are elevated in soils especially within 300 mm from the pole (Chirenjea et al., 2003).

Mobility of the metals within soils is affected by many factors. It is affected not only by soil pH but also the cation exchange capacity of soil. Indefinitely, both are affected by organic matter concentration in soil. A soil with lower pH will leach the metals while a soil with higher cation
exchange capacity will improve cationic holding capacity of soil. Soil with higher organic matter will reduce pH and at the same time increase cationic exchange capacity.

Arsenic is more mobile than the other two CCA components and it can leach more than 8 times compared to Copper elements from a newly preserved wood (Bergholm, 1990; Rahman et al., 2004; Stilwell et al., 2006 and Mercer et al., 2012).

2.2.2 Factors affecting leaching

There is no doubt that CCA components leached out from wood surface to the soil surrounding treated wood structures based on research by Lebow, (1996), Cooper and Ung, (1997), Solo-Gabriele et al., (2000), and Townsend et al., (2001). Leaching of CCA from wood is dependent on many factors, both internal (wood elements) and external (weather and soil properties) (Chirenjea et al., 2003).

Different CCA formulation used is dependent on where and how it is used and the application method. These can affect the leaching of metals to the environment. At the same time, retention plays a big role in determining leaching of CCA as different retention factors like type of timber, species of timber, intended usage, and the service life expectancy can determine leaching rate of arsenic, chromium and copper (Chirenjea et al., 2003).
Another factor that affects leaching rate is physical-chemical properties of immediate soil. Metal concentrations are distributed according to soil characteristics and soil physical-chemical properties influence the leaching of CCA components (Townsend et al., 2003). Soils with high organic matter (OM) are known to have lower pH due to organic acids. Solubility of the metals are pH dependent and increases with decreasing pH level (Bergholm, 1990), thus higher organic matter in soil will lead to increase leaching of CCA components. Organic matter is also known to form chelate complexes with heavy metal. These organic ligands can react with CCA metal components and thus retain them on the surface of soil where most organic matter occurs, supporting findings of Townsend et al. (2003) and Morrell and Huffman (2004) showing greatest concentrations of CCA metal elements (Cr, As, and Cu) occurring at the surface.

Weathering of treated wood can cause elevated levels of Arsenic, Chromium and Copper in the surrounding soil of CCA-treated timber. Increased precipitation and intensity of sunlight will degrade the timber faster and thus increases the effect of weathering on the integrity of CCA component fixation in the wood. Soil moisture is shown to have impact on leaching whereby wetter soil was more conducive to leaching of CCA components (Cooper, 1994; Mercer et al., 2012). Furthermore, the metals do leached over time when exposed to water (Lebow, 1996; Hingston et al., 2001). As a matter of fact, precipitation over treated timber and water logged soil will dissolve the CCA components and leached into the environment (Townsend et al., 2003). In fact, with the wet and humid tropical climate of Sarawak, Malaysia, elevated precipitation levels is expected to play a big role in leaching of CCA components to the soil environment.
Leaching is also affected by fixation rate of the components in wood and improperly fixed components leaches out readily compared to properly fixed components. CCA components are termed “fixed” when the chromium (VI) species is reduced to chromium (III) by reaction with the wood components (Cooper et al., 2004). This reduction by wood components lead to precipitation and stabilization of copper and arsenic and is responsible for the low leachability of metals and its efficiency in protecting wood from decay (Cooper et al., 2004). Chemical reaction that takes place in wood preservation of CCA may not be as “fixed” permanently as believed when researchers determined that CCA components migrate from wood over time (Cooper, 1994; Morrell and Huffman, 2004). Leaching of CCA is significant after treatment when unfixed components leached to the environment (Morrell and Huffman, 2004). Additionally, Salim et al., (2012) conducted experiments to investigate this relationship with Malaysian hardwood timber and found that high amounts of CCA remain as unfixed state resulting in higher leaching of metals in the leachate especially Ramin and Geronggang. Additionally, different wood species differs in its chemistry (eg: pH, extractives) and anatomy (eg: permeability, cell wall thickness, density) resulting in difference in leaching (Cockroft and Laidlaw, 1978). More permeable species are easier to be treated and at the same time, a tendency to leach at a higher rate (Cockroft and Laidlaw, 1978).

Cooper and Ung (1992) reported that CCA retention in wood has little influence on fixation rate. Additionally, no significant relationship between leaching and retention has been implied although leaching percentage and concentration is affected by initial CCA salt loading (Cooper, 1994). Furthermore, incomplete fixation may occur due to presence of extractives which can
interfere in the fixation process of CCA components (Dahlgren, 1975; Kennedy and Palmer, 1994; Cooper et al., 1997; Stevanovic-Janezic et al., 2000).

In fact, Salim et al., (2012) detected that Malaysian tropical hardwood (i.e: *Cratoxylum* sp., *Gonystilus* sp., *Calophyllum* sp., *Litsea* sp., *Shorea acuminate*, and *Dillenia* sp.) had recorded higher rate of unfixed CCA components and higher rate of leaching especially arsenic when compared to Radiata Pine wood. Regardless of fixation time, high incidence of incomplete fixation of CCA especially *Geronggang* (*Cratoxylum* spp.) and *Ramin* (*Gonystilus* spp.) with poor CCA fixation characteristics was confirmed by leaching tests showing high losses of CCA components (Salim et al., 2012).

2.3 Effects of leached CCA to the environment

Human activities should be taken into serious consideration on how it had greatly contributed to heavy metal pollution to the environment. Heavy metals occurring at high doses and elevated exposure can have adverse impacts on the environment and also the health. Heavy metals stay in the environment and is not easily removed. All these pollutants can bioaccumulate in the system which will concentrate and biomagnifications can heavily affect the high tiered consumer in the food chain. CCA active components; arsenic, chromium and copper are categorized as heavy metals being partially responsible for environmental pollution.
Leaching of chromium, copper and arsenic from treated timber causes elevated levels of heavy metal to the surrounding environment. Usage of CCA treated timbers in large quantities can pollute the environment and may even have health repercussions to public exposed to them. Treated timbers planted in the soil can pollute the soil environment to several times its background concentration. Arsenic as one of the components of CCA is of main environmental concern. This is because arsenic is highly toxic to living organisms. Consequently, plants can assimilate arsenic within its own cells and when eaten by animals will bioaccumulate into the food chain. In fact, Rahman et al., (2004) and Stilwell et al., (2006) concluded that growing vegetables in soils near CCA treated wood or CCA contaminated soil may pose increased risk of exposure to arsenic.

CCA treated wood are highly durable and at the end of its service life, are disposed off in landfills or burned. Such disposal posed problems due to potential release of heavy metals to the environment (Hasan et al., 2010; Coudert et al., 2012). Contaminated soils also require consideration as this has led to costly soil remediation measures in parts of Europe and North America to render the area as decontaminated.

Effects of heavy metals on human health are quite extensively studied by organizations like World Health Organization (WHO). The effect ranges from a minor headache to death depending on the exposure to the metallic species. The current situation of the world is that we are increasingly exposed to heavy metal pollutants either through breathing in air, drinking water or just by eating food items available in the market.
Inorganic arsenic is widely distributed and present in groundwater used for drinking in many countries whereas organic arsenic are found to be present in the food chain thus may risk human exposure (Järup, 2003). Levels in soil usually ranged 1 to 40 mg/kg (Järup, 2003), but anthropogenic activities may elevate the levels in the soils. Inorganic arsenic is acutely toxic and adequate intake can lead to severe cardiovascular and gastrointestinal symptoms and disturbance of central nervous systems that will lead to death (Järup, 2003). Besides that, via exposure to arsenic in drinking water with concentration of more than 100 μg/L are known to cause cancer in lungs, kidney, bladder and skin (Järup, 2003).

Other than arsenic, another potential hazard is chromium. Toxicity of both chromium (III) and chromium (VI) are known. Toxicity of chromium (III) is less than its more mobile and highly oxidized chromium (VI) due to its difference in state of stability between the Cr species. Chromium (VI) is a well known carcinogenic agent (Barceloux and Barceloux, 1999) whereas, high concentration of chromium(III) in the cell can lead to DNA damage as well (Eastmond et al., 2008) even though it is lower in toxicity compared to chromium (VI).

Overall, such dangers posed to the soil environment as a response to leaching of CCA preservatives from treated wood use, is perhaps serious after several years of exposure of treated wood. However, dangers posed by short term exposures should also be considered in future studies.
3.0 MATERIALS AND METHODS

3.1 Pole samples

The poles are treated with chromated copper arsenate (CCA) Tanalith-C® (35% Cu as CuSO₄·5H₂O, 45% Cr as K₂Cr₂O₇ and 20% As as As₂O₅·2H₂O) using Full Cell (Bethell) process about 30 years ago at Timber Research and Technical Training Centre. Briefly, the poles are put in a CCA pressure treatment plant, vacuumed for one hour and flooded with the preservatives. The chamber is then pressurized at 200 psi (14 bars) for two hours to treat till refusal when CCA salts are no longer able to be absorbed by the wood before the chamber is vacuum drained to remove the leftover preservatives for half an hour.

The 8 feet in length poles are placed 3 x 4 metres perpendicular and 2.5 metres at 45 degrees angular as shown on Figure 1. The poles are planted two feet below ground since early 1980 and 1981 at Timber Research and Technical Training Centre (TRTTC).

![Figure 1: Planting distance of the poles.](image)
Timber Research and Technical Training Centre (TRTTC) has been leading research on wood related technologies since its inception on 21st August 1971 in Sarawak, Malaysia. As it is situated near the equator, the poles are influenced by severe weathering due to direct exposure to the sun rays, high humidity and high precipitation of the tropical climate.

3.2 Soil

3.2.1 Sampling of soil

Soil sampling methods is adapted from the methods used by Morrell and Huffman (2004). Soil auger was used to collect soil samples around each pole based on horizontal distance away and vertical depth at each selected horizontal plane. Soil was sampled horizontally at distance 0 mm, 150 mm and 300 mm away from the post and at vertical depth of 0-50 mm, 150-200 mm and 300-350 mm at each corresponding sampling point. Background metal levels in soils were accessed by taking soil samples some distance away from the test site and at similar vertical depth levels. All the samples were oven dried at 105°C for five hours. Then, dried samples are crushed with mortar and pestle before sieved using 0.425 mm sieve to obtain homogeneous sample for extraction and analysis.

3.2.2 Soil metal extraction (Aqua regia digestion).

Two gram of soil was put into 50 mL beaker. Next, 20 mL of aqua regia (AR) solution (5 mL concentrated nitric acid (HNO₃) [69-70% m/m] and 15 mL concentrated hydrochloric acid (HCl) [37% m/m]) was added and heated to 130°C for two hours. Then, the contents were cooled after
rinsing the beaker with distilled water. After cooling, 1 mL concentrated nitric acid (HNO₃) [69-70% m/m] and 20 mL of distilled water was added before reheating at 130°C. After heating for one hour, the contents were cooled at room temperature and the solution filtered using Whatman No 42 filter paper. The filtrate was transferred into 100 mL volumetric flask and distilled water added to make up the mark at 100 mL.

3.2.3 Soil pH

The pH of soil was tested only on the control soil sample to represent the soil in the area as the size of sampling field is less than 1 hectare. The soil was oven dried at 105°C for five hours before grinding and sieved through a 2 mm sieve. Five grams of soil and 25 mL of distilled water was put into a beaker. The beaker is agitated and mixed evenly using a magnetic stirrer for 2 hours. Next, the soil-water mixture’s pH was measured using a pH meter.

3.3 Wood

3.3.1 Sampling and preparation

Wood samples approximately 12 mm in depth from the surface were removed from the pole at 1.3 m above ground and at 0-20 cm below ground level. After that, the wood samples were pulverized using a mill and sieved using 0.425 mm test sieve. Then, the woodmeal was mixed evenly and oven dried at 105 °C for 24 hours.
3.3.2 Metal extraction from wood (Copper, Chromium and Arsenic)

Extraction of wood preservative components is adapted from AS/NZS 1605 Standard (AS/NZS 2000). Two gram of woodmeal was transferred into a 250 mL conical flask. Then, 40 mL of 2.5M sulphuric acid (H$_2$SO$_4$) solution and 8 mL of 30% hydrogen peroxide (H$_2$O$_2$) solution were added. The mixture was heated at 75°C in a water bath for 30 minutes with occasional agitation of the mixture. After that, 80 mL of distilled water was added and reheated until ceasing of evolution of bubbles signaling the full degradation of hydrogen peroxide. The mixture was filtered using Whatman No 42 filter paper and transferred into a 250 mL volumetric flask. Next, 20 mL of 3% sodium sulphate (Na$_2$SO$_4$) solution was added and distilled water used to top up to the 250 mL mark.

3.3.3 Density of wood

Methods for the measurement of wood density were adopted from the ASTM D 2395-93 (ASTM, 2000). Briefly, a single wood sample (uneven surface) was obtained from each pole aboveground and oven dried at 105 °C for 24 hours and weighed oven dry. Next, a beaker was filled ¾ full with distilled water and tarred before the test. The wood sample was then immersed in the water by using a pin without touching the corner and the bottom of the beaker. The weight of water displaced by the immersed sample was obtained which is a measure of the wood volume (cm$^3$).

- Volume (cm$^3$) of wood piece = Immersed specimen (g) (density of water is 1.00 g/ml).
• Density of treated wood = \( \frac{\text{dry weight of the wood}}{\text{volume of wood}} \) and expressed in kg/m\(^3\).

• Initial wood density = Density of treated wood – Retention value of CCA salt and expressed in kg/m\(^3\).

3.4 Analysis and measurement of metal levels (Copper, Chromium and Arsenic)

The prepared extracts were then analyzed using Flame Atomic Absorption Spectrometry (Thermo Fisher’s iCE 3000 Series AA). Results were recalculated to reflect the concentration of the metals in the samples and then subjected to statistical analysis using SPSS Statistical software Ver. 17.0. Flame AAS detection limit of Arsenic is at 0.12 ppm, Copper at 0.0045 ppm and Chromium at 0.0054 ppm.

Flame AAS is insensitive to Arsenic and with the extraction methods used above can only accurately detect arsenic concentrations up to 6.0000 ppm and above.

3.4.1 Retention value of CCA salts in wood

In order to calculate the weight of retained CCA salts in wood, metal concentrations obtained from AAS were expressed in terms of their respective metal salts by multiplying with the following factors:
Copper as copper sulphate (CuSO₄·5H₂O) = 3.93
Chromium as potassium dichromate (K₂Cr₂O₇) = 2.83
Arsenic as arsenic pentoxide (As₂O₅·2H₂O) = 1.77

The dry salt retention in treated wood was expressed as kg/m³ as the density of the wood was known from earlier test. The retention of CCA salt in oven dried wood is given by:

\[
\frac{(A + B + C) \text{mg/kg}}{1,000,000} \times D \text{(kg/m}^3)\]

Where;

A = Concentration of Arsenic salt
B = Concentration of Chromium salt
C = Concentration of Copper salt
D = Initial density of wood